Half-Integer Shapiro Steps at the 0-π Crossover of a Ferromagnetic Josephson Junction

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We investigate the current-phase relation of S/F/S junctions near the crossover between the 0 and the π ground states. We use Nb/CuNi/Nb junctions where this crossover is driven both by thickness and temperature. For a certain thickness a nonzero minimum of critical current is observed at the crossover temperature. We analyze this residual supercurrent by applying a high frequency excitation and observe the formation of half-integer Shapiro steps. We attribute these fractional steps to a doubling of the Josephson frequency due to a $\sin(2\phi)$ current-phase relation. This phase dependence is explained by the splitting of the energy levels in the ferromagnetic exchange field.

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The current-phase relation of ballistic and diffusive S/N/S junctions is predicted to be strongly nonsinusoidal at zero temperature in contrast to that of tunnel junctions. This is due to a different conductivity mechanism involving Andreev bound states created in the normal metal (N) by the superconductors (S). These states are sensitive to the superconducting phase difference ϕ and carry the supercurrent I_S. In S/F/S junctions the current-phase relation is strongly distorted by the exchange field of the ferromagnet (F) and can even be *reversed* leading to the famous π state [1]. The microscopic mechanism responsible for this *negative* supercurrent can be intuitively explained in the clean limit where the Andreev spectrum is discrete [2–4]. The two spin configurations of each bound state are indeed split by the ferromagnetic exchange energy. When the first bound state is shifted from finite energy to zero energy (at $\phi = 0$), the direction of the total supercurrent given by the lowest level (for $\phi > 0$) is *negative* (instead of positive). In this case the ground state is at $\phi = \pi$.

In this Letter we consider the situation where the exchange energy E_{ex} is half that of the π junction described above for the same thickness d. In this case the Andreev spectrum of a ballistic junction contains equidistant states twice closer than usual [Fig. 1(a)]. As a result the supercurrent is π periodic in phase with a sawtooth shape at zero temperature [Fig. 1(b)], and the ground states $\phi = 0$ and $\phi = \pi$ are degenerate [5]. The current-phase relation becomes more rounded in the diffusive regime [Fig. 1(c)] where the discrete spectrum is replaced by a continuous density of Andreev states [6]. At this 0-π crossover the current-phase relation contains a dominant $\sin(2\phi)$ component and the critical current presents a nonzero minimum with respect to thickness or exchange energy variations. Experimentally the critical current of S/F/S junctions at the crossover is, however, so small that it was always assumed to vanish completely. In Nb/CuNi/Nb junctions [4,7] this behavior could be related to the strong decoherence of the magnetic alloy and in Nb/PdNi/Al_2O_3/Nb junctions [8] to the presence of the tunnel barrier.

In this Letter we report the first observation of a small nonzero critical current at the 0-π crossover of a Nb/CuNi/Nb junction and show that the corresponding current-phase relation has the expected $\sin(2\phi)$ dependence. An evidence of such a relation could be obtained with a two junction superconducting loop: if one of them has a $\sin(2\phi)$ relation, the interference pattern under magnetic field should have maxima both at integral and half-integral flux quanta [5], whereas the maxima occur only at the integral (half-integral) values if this junction is fully in the 0 (π) state [9,10]. However, we chose to analyze the current-phase relation by studying the dynamic behavior of a single junction. Up to now only the equilibrium properties of the S/F/S junctions have been theoretically and experimentally investigated. In this

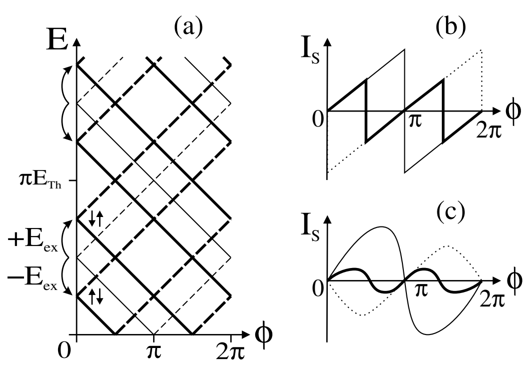

FIG. 1. (a) Energy of Andreev levels versus phase difference in a one-dimensional ballistic junction ($E_{Th} = \hbar v_F/d \ll \Delta$) [4]. Each state of the normal case (thin lines) is split by the exchange energy E_{ex} of the ferromagnet (thick lines). The states in solid (dashed) lines carry positive (negative) currents. (b) Current-phase relation for this model at zero temperature in the normal case (thin line), in π junctions (dotted line), and at the crossover (thick line). (c) Current-phase relation in diffusive regime for the same situations.

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Letter we present the first study of the finite voltage behavior under high frequency excitation to reveal the harmonics of the supercurrent. In the junction with a nonzero critical current at the crossover we observed half-integer Shapiro steps attributed to a $\sin(2\phi)$ current-phase relation. This phase dependence reveals the level splitting induced by the ferromagnetic exchange field.

Our junctions are $Nb/Cu_{52}Ni_{48}/Nb$ trilayers deposited *in situ* and patterned by photolithography. The copper-nickel alloy has a Curie temperature as small as 20 K. Details of these junctions and of the experimental setup have been reported previously (Ref. [4]). In this Letter we analyze the behavior of two junctions with copper-nickel thicknesses equal to 17 and 19 nm (the normal resistances R_N are equal to 0.12 and 0.13 mΩ, respectively). The temperature dependence of their critical current I_C is shown in Fig. 2. These unusual behaviors are explained by the spectral supercurrent density of diffusive S/F/S junctions [4,6]. The ground state switches from $\phi = \pi$ to $\phi = 0$, and the critical current presents a deep minimum at the crossover temperature T^*. In our set of samples [4], only the 17, 18, and 19 nm thick junctions have this 0-π crossover, at T^* equal to 1.12, 4.53, and 5.36 K, respectively. Junctions thinner and thicker have, respectively, a 0 and a π ground state at all temperatures.

An expanded view of the crossover region reveals a finite critical current of 4 μA at T^* for the 17 nm thick junction. We show in the following that this supercurrent has a $\sin(2\phi)$ current-phase relation. With a 0-π crossover close to zero temperature, this junction is indeed very close to the optimum situation where the ratio E_{ex}/E_{Th} gives a doubled periodicity of Andreev levels. The vanishing critical current obtained for the 19 nm (and 18 nm) thick junction may be explained by the larger thickness giving a more stable π state at zero temperature. It could also be related to the higher value of T^*, since the $\sin(2\phi)$ component is expected to become much smaller when the temperature is high (of the order of the critical temperature T_c of the electrodes) [5].

The current-phase relation is analyzed by sending an alternative current at 800 kHz when measuring the direct voltage-current curve (we use a rather low frequency because the characteristic voltage $R_N I_C$ is in the nanovolt range as a consequence of the very small critical current). Constant voltage Shapiro steps appear due to the synchronization of the Josephson oscillations on the applied excitation. The curves obtained for the 19 nm thick junction are shown in Fig. 3. As expected the steps appear at voltages equal to integral multiples of $\Phi_0 f = 1.6$ nV ($\Phi_0 = h/2e$). They are similar above and below the crossover temperature at 5.36 K where the critical current and the steps disappear simultaneously.

The curves obtained for the 17 nm thick junction with the same excitation amplitude and frequency are shown in Fig. 4. The integer Shapiro steps are present at all temperatures, including 1.12 K where the critical current is minimum but nonzero. The new result is that two additional steps appear at voltages $(1/2)\Phi_0 f$ and $(3/2)\Phi_0 f$ at this crossover temperature. Small features at the same voltages are also present at 1.10 K, but almost nothing is visible at 1.07 K. These half-integer steps reveal the existence of supercurrent oscillations at frequency $2(V/\Phi_0)$ which synchronize to the excitation at frequency f producing steps at voltage multiples of $(1/2)\Phi_0 f$. This doubling of the Josephson frequency is the consequence of the $\sin(2\phi)$ current-phase relation expected at the 0-π crossover of S/F/S junctions.

Ideally, the $\sin(2\phi)$ component should dominate for this thickness of junction at all temperatures $T \ll T_c$. The fact that these half-integer steps are visible only close

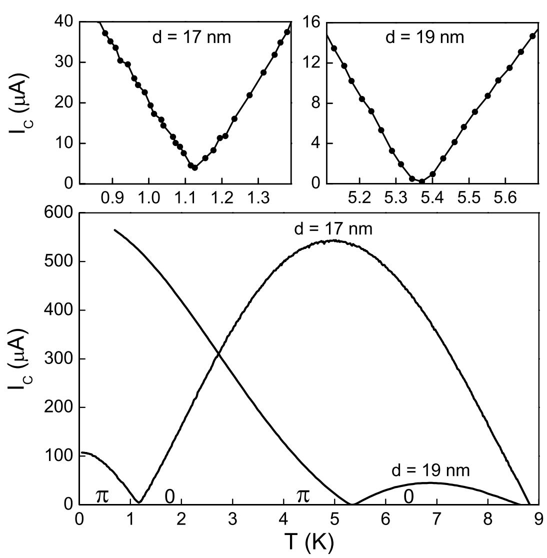

FIG. 2. Temperature dependence of the critical current for two $Nb/Cu_{52}Ni_{48}/Nb$ junctions. The upper graphs are expanded views near the crossovers between the 0 and π states: the critical current has a nonzero (zero) minimum for the 17 nm (19 nm) thick junction.

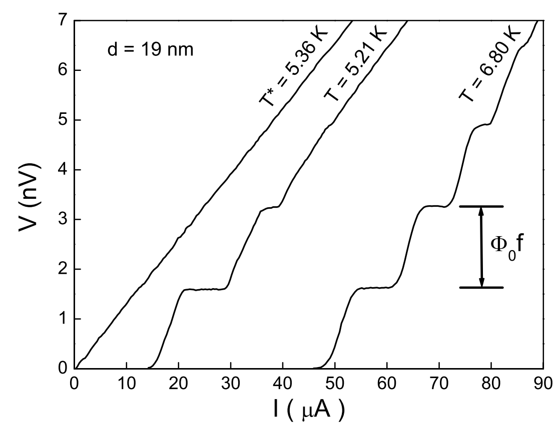

FIG. 3. Integer Shapiro steps in the voltage-current curve of a 19 nm thick junction with an excitation at 800 kHz (amplitude about 18 μA). The steps disappear at the 0-π crossover temperature T^*. Curves at 5.21 and 6.80 K are shifted by 10 and 20 μA for clarity.

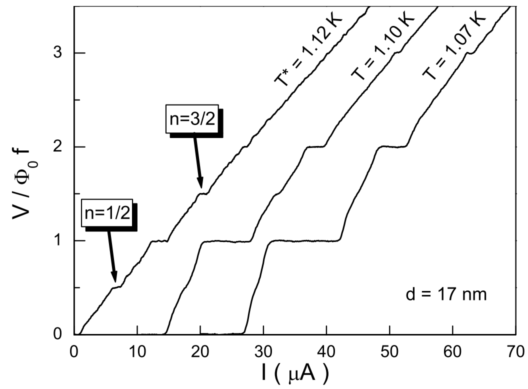

FIG. 4. Shapiro steps in the voltage-current curve of a 17 nm thick junction with an excitation at 800 kHz (amplitude about 18 μA). Half-integer steps ($n = 1/2$ and $n = 3/2$) appear at the 0-π crossover temperature T^*. Curves at 1.10 and 1.07 K are shifted by 10 and 20 μA for clarity.

to T^* indicates that the phase dependence departs very slightly from $\sin(\phi)$. The $\sin(2\phi)$ component dominates only when the large $\sin(\phi)$ component cancels to change its sign (i.e., when the temperature induces the compensation of the negative and positive currents at $\phi = \pi/2$). To the first order near T^*, the supercurrent can be qualitatively described by the relation $I_S(T) = \frac{T-T^*}{T^*} I_1 \sin(\phi) + I_2 \sin(2\phi)$. The fact that $I_1 \gg I_2$ may be related to the strong decoherence in the magnetic alloy (also responsible for the huge reduction of the critical currents when compared with theoretical predictions [4]). If the superconducting correlations are small enough, the equations can be linearized and give indeed a $\sin(\phi)$ relation regardless of the exchange energy. A theoretical study of diffusive S/F/S junctions taking into account the spin-flip scattering would be required to analyze this behavior in more detail.

In this paragraph we present other kinds of junctions where fractional steps have been observed in the past in order to show the specific origin of the half-integer steps observed in our junctions. In a current biased junction the supercurrent oscillations at finite voltage are strongly nonsinusoidal and the harmonics could synchronize on the excitation leading to fractional steps. However, simulations have shown that this mechanism does not occur in the resistively shunted junction (RSJ) model with a $\sin(\phi)$ current-phase relation [11]. Fractional steps can appear if a large capacitance or inductance is present, but this is not the case in our nonhysteretic junctions because of their large conductance and small critical current. Fractional steps can also appear in the case of a nonsinusoidal relation which is in fact expected from theory in any kind of weak link at sufficiently low temperature [12–14]. They were observed in superconducting microbridges and point contacts but are more difficult to observe in diffusive S/N/S junctions: for junctions with thick N layers compared to the coherence length, the low temperature regime is hard to reach; and for shorter junctions with a sandwich structure, the large cross sections imply large critical currents that cannot be measured in the low temperature regime (this is, however, possible in our case thanks to the strong decoherence by spin-flip scattering). Fractional steps have been observed at high temperature in long and wide diffusive S/N/S junctions in sandwich geometries and were attributed to a synchronization of the vortex flow [15]. This flux-flow cannot happen in our junctions since the Josephson penetration length λ_J is much larger than the junction width. Fractional steps have also been observed in planar S/N/S junctions at high temperature where the critical current has vanished [16,17]. These steps may involve dynamical and nonequilibrium effects acting on the phase-coherent contribution to the resistance.

In contrast to these experiments, our half-integer steps are visible only at the crossover, at low temperature, and with a finite critical current. The interpretation of these half-integer steps as a consequence of a current-phase relation with a $\sin(2\phi)$ dependence seems therefore reasonable. Since the transparency deduced from the normal state resistance is good [4], this phase dependence has to be related to an Andreev conduction mechanism (as opposed to tunneling) and is explained by the doubled periodicity of Andreev levels at the 0-π crossover.

We now investigate in more detail the harmonic composition of the supercurrent in the 17 nm thick junction at 1.12 K. For this purpose we measure the voltage-current curve for different excitation amplitudes [Fig. 5(a)] and compare quantitatively the width of the Shapiro steps to the prediction of the RSJ model for non-$\sin(\phi)$ current-phase relations [Fig. 5(b)]. During the experiment we measured only the relative amplitudes of the alternative current, but for a quantitative comparison we need the absolute values I_{ac}. Since no half-integer step is observed at 1.07 K we can assume a $\sin(\phi)$ relation and adjust the step widths to the result of the RSJ model, yielding an excitation amplitude of 18 μA. In this case the normalized half-width I_n/I_C (the full width is $2I_n$) is equal to the Bessel function $J_n(a)$ when $R_N I_C < \Phi_0 f$ and where $a = R_N I_{ac}/\Phi_0 f$ [18]. Numerical simulations of the voltage-current curves have been performed using a $\sin(2\phi)$ current-phase relation and the dependence of the step widths with the excitation amplitude has been extracted. As expected the steps are proportional to the Bessel functions $J_{2n}(2a)$. In particular, the $n = 1/2$ step has the same behavior as the $n = 1$ step of a usual junction with an excitation 2 times larger. This $n = 1/2$ step is not a subharmonic step but the fundamental step for a double Josephson frequency. Experimentally the width of this $n = 1/2$ step first increases and then decreases with the excitation amplitude in agreement with the oscillating behavior of the Bessel functions. However, its width is significantly smaller than expected for a pure $\sin(2\phi)$ current-phase relation [Fig. 5(b), solid line). This difference may be the consequence of a residual $\sin(\phi)$ component in the supercurrent. Numerical simulations

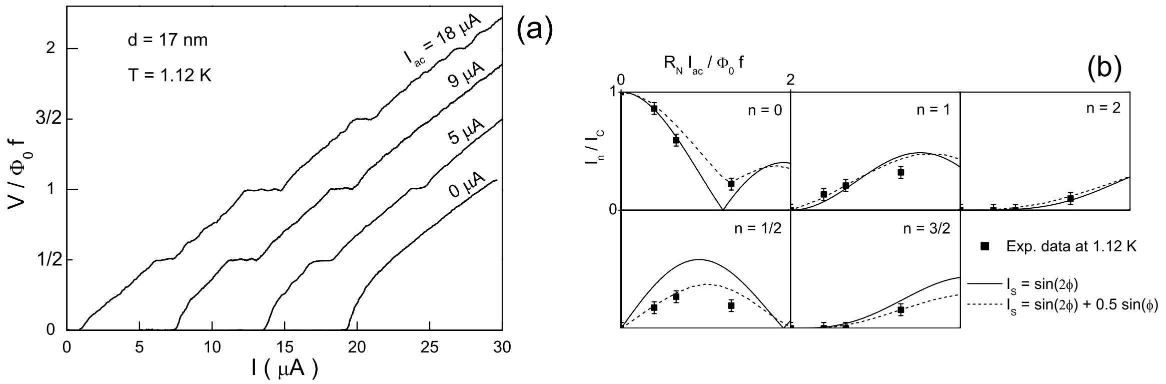

FIG. 5. (a) Shapiro steps in the voltage-current curves of the 17 nm thick junction at 1.12 K with an excitation at 800 kHz. Excitation amplitudes are about 18, 9, 5, and 0 μA. The respective curves are shifted by 0, 5, 10, and 15 μA for clarity. (b) Width of the integer and half-integer steps versus normalized amplitude of the alternative current. Experimental values (symbols) are compared with numerical simulations for two current-phase relations containing a dominant $\sin(2\phi)$ component (lines). All graphs have the same axis and scales.

have been performed to calculate the step widths for current-phase relations containing the two phase dependencies with different ratios. The best fit to the experimental data is obtained when the amplitude of the $\sin(\phi)$ is half that of the $\sin(2\phi)$ equal to 3.2 μA (dashed line). This analysis indicates that the crossover temperature, where the $\sin(\phi)$ component should disappear completely, is not exactly 1.12 K. From the slope of the critical current versus temperature (Fig. 2) and the amplitude of the residual $\sin(\phi)$ component (1.6 μA) we can, however, estimate the actual crossover to be only 12 mK above or below this temperature.

In summary we studied the finite voltage behavior of S/F/S junctions under high frequency excitation and observed half-integer Shapiro steps at the temperature of a 0-π crossover where the critical current is nonzero. These steps reveal the $\sin(2\phi)$ dependence of the current-phase relation which is explained by the specific level splitting realized at the crossover. This unusual relation changes rapidly for a $\sin(\phi)$ dependence when one moves away from the crossover temperature.

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[1] A. I. Buzdin, L. N. Bulaevskiĭ, and S. V. Panyukov, Pis'ma Zh. Eksp. Teor. Fiz. **35**, 147 (1982) [JETP Lett. **35**, 178 (1982)].
[2] L. Dobrosavljević-Grujić, R. Zikić, and Z. Radović, Physica (Amsterdam) **331C**, 254 (2000).
[3] A. A. Golubov, M. Y. Kupriyanov, and Y. V. Fominov, Pis'ma Zh. Eksp. Teor. Fiz. **75**, 709 (2002) [JETP Lett. **75**, 588 (2002)].
[4] H. Sellier, C. Baraduc, F. Lefloch, and R. Calemczuk, Phys. Rev. B **68**, 054531 (2003).
[5] Z. Radović, L. Dobrosavljević-Grujić, and B. Vujičić, Phys. Rev. B **63**, 214512 (2001).
[6] T. T. Heikkilä, F. K. Wilhelm, and G. Schön, Europhys. Lett. **51**, 434 (2000).
[7] V. V. Ryazanov, V. A. Oboznov, A. Y. Rusanov, A. V. Veretennikov, A. A. Golubov, and J. Aarts, Phys. Rev. Lett. **86**, 2427 (2001).
[8] T. Kontos, M. Aprili, J. Lesueur, F. Genêt, B. Stephanidis, and R. Boursier, Phys. Rev. Lett. **89**, 137007 (2002).
[9] V. V. Ryazanov, V. A. Oboznov, A. V. Veretennikov, and A. Y. Rusanov, Phys. Rev. B **65**, 020501 (2001).
[10] W. Guichard, M. Aprili, O. Bourgeois, T. Kontos, J. Lesueur, and P. Gandit, Phys. Rev. Lett. **90**, 167001 (2003).
[11] C. A. Hamilton and E. G. Johnson, Phys. Lett. A **41**, 393 (1972).
[12] J. Bardeen and J. L. Johnson, Phys. Rev. B **5**, 72 (1972).
[13] I. O. Kulik and A. N. Omelyanchuk, Zh. Eksp. Teor. Fiz. Pis'ma Red. **21**, 216 (1975) [JETP Lett. **21**, 96 (1975)].
[14] F. K. Wilhelm, G. Schön, and A. D. Zaikin, Phys. Rev. Lett. **81**, 1682 (1998).
[15] J. Clarke, Phys. Rev. Lett. **21**, 1566 (1968).
[16] K. W. Lehnert, N. Argaman, H.-R. Blank, K. C. Wong, S. J. Allen, E. L. Hu, and H. Kroemer, Phys. Rev. Lett. **82**, 1265 (1999).
[17] P. Dubos, H. Courtois, O. Buisson, and B. Pannetier, Phys. Rev. Lett. **87**, 206801 (2001).
[18] K. K. Likharev, Rev. Mod. Phys. **51**, 101 (1979).